STATUS OF THE BACK-END OPTIONAL ADVANCED RESEARCH REACTOR FUEL DEVELOPMENT IN KOREA

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ABSTRACT

U-Mo fuel development has been carried out for a reactor upgrade of HANARO and the back-end option in Korea. The 2nd irradiation test of the U-Mo dispersion rod fuels is underway in HANARO in order to find the optimum uranium loading density and to investigate the applicability of the monolithic U-Mo ring fuel as well as other parameters such as particle size and cladding surface-treatment. The optical observation using an immersion camera showed that the cladding surfaces of the two U_3Si and U-Mo fuels with a high power rate changed in to the darker color, which is not as severe as those of the driving fuels in HANARO. Presumably it would be acceptable. The other fuels were observed as maintaining their initial good conditions. In connection with monolithic U-Mo fuel development, some achievements such as preliminary U-Mo tube production by a continuous casting process and a successful U-Mo foil production using a roll casting process have been obtained. In addition, some investigation on the surface-treatment of multiplayer coating and Zr sputtering coating has showed the possibility of eliminating the problem of a temperature rise due to the corrosion layer formation having quite a low conductivity. The next irradiation test will aim mainly at the qualification of the U-Mo dispersion fuel for HANARO around the end of next year. In the 3rd irradiation fuel buddle, some fuels related to the basic investigation tests for the monolithic U-Mo fuel and surface-treatment for anticorrosion will be loaded.

1. Introduction

In connection with the back-end option and the reactor upgrade by applying the higher U-density, the qualification of the U-Mo dispersion rod fuel has been carried out[1]. The first irradiation test for the U-Mo dispersion rod fuels revealed that the fuels with a U-loading of 6.0 g-U/cc weren't acceptable due to the complete interaction between the U-Mo particles and Al matrix induced[2]. The most strongly affecting parameter was concluded to be the fuel temperature. In the 2nd irradiation test lower U-density fuels were targeted in order to reduce the temperature. The fuel meat diameters were designed to be 5.49 mm and 6.35 mm for 4.5 g-U/cc and 4.0 g-U/cc, respectively. One U-Mo monolithic ring (^{OD}6.2 mm × ^{t}0.7 mm) with Al cladding was loaded in order to avoid the severe interaction problem. In addition, Two different particle size fuels, two surface treated fuels with pre-oxidation and Ni coating, one fuel rod added with the poison material of Er_2O_3, A fuel rod dispersed with U-9wt.%Mo powder, and a U_3Si dispersion fuel rod were loaded. The fuel assembly with the above 10 fuel rods has been irradiated in HAHARO since January 9, 2003 for the target burnup of 60 at.%. During the maintenance period of HANARO some observations were made on the cladding surfaces of the irradiating fuel rods by using an immersion camera. The maximum burnup for the U-Mo dispersion fuel rods as of July 30 was calculated at about 25 at.%.

The main purpose of the 2nd irradiation test for the qualification is to find the optimum uranium loading density of the U-Mo dispersion rod type fuel. For the qualification it is planned that real-size U-Mo dispersion fuel rods are irradiation-tested at the selected uranium density in the 3rd test.

Separately efforts have been made for the fabrication technology development of the monolithic U-Mo thin tube, ring, and foil as well as the cladding surface treatment technologies such as multi-layer coating and Zr coating by sputtering[3][4].

In this paper the optical observation for the fuel claddings under the 2nd irradiation test, the progress on the fabrication technology development of the U-Mo monolithic fuel, and the plans of the next irradiation test related to the U-Mo fuel qualification in Korea are reported

2. The status of the 2nd irradiation test of U-Mo dispersions and monolithic fuels

2-1. The fuels loaded in the 2nd irradiation test

The 10 fuel rods loaded in the 2nd irradiation test are shown in Table 1. The specification is the same as the HANARO fuel except the fuel meat length. All the fuel powders of U-Mo and U_3Si were prepared by the atomization process. The particle size distributions of the different size fuel meats are represented in the Table 2.

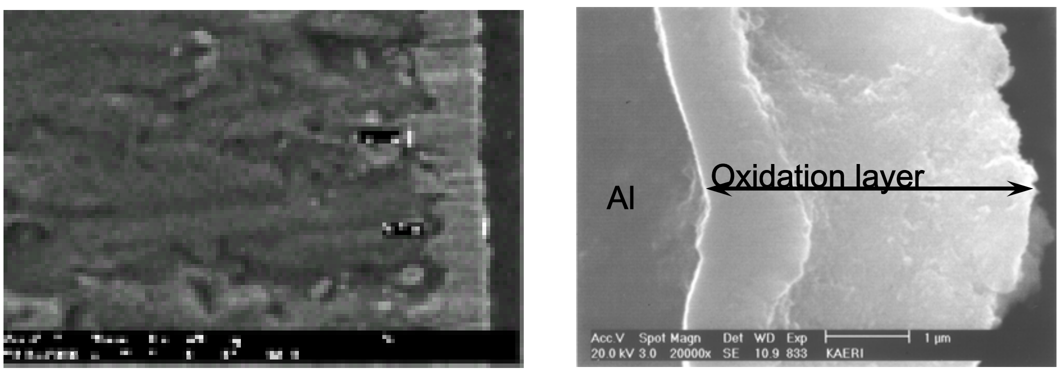
Table 1. The fuels loaded in the 2nd irradiation test

Serial No.	Fuel Material	Loading (g-U/cc)	Diameter (mm)	Length (mm)	Number	Remarks
1	U-7Mo	4.5	5.49	360	1	High density
2	U-9Mo	4.5	5.49	360	1	High Mo
3	U-7Mo	4.0	6.35	360	1	Low density
4	U-7Mo	4.0	6.35	110	1	Larger particles
5	U-7Mo	4.0	6.35	110	1	Smaller Particles
6	U3Si	4.0	6.35	210	1	Reference
7	U-7Mo	4.5	5.49	210	1	Poison
8	U-7Mo	Pure	$^{OD}6.2\times{}^{t}0.7$	3.25	1	Ring
9	U-7Mo	4.0	6.35	360	1	Ni coating
10	U-7Mo	4.0	6.35	360	1	Pre-oxidation

Table 2. Particle size distributions of two different particle size fuels.

	Smaller particle fuel		Larger particle fuel	
Size distribution	38～45 ㎛	11.1%	53～63 ㎛	36.3%
	45～53 ㎛	59.2%	63～75 ㎛	40.0%
	53～63 ㎛	29.7%	75～90 ㎛	19.3%
			90～106 ㎛	4.4%

In order to avoid the temperature increase by the corrosion layer of the cladding, two surface-treated fuels were loaded in the 2nd irradiation test. Ni was coated on the cladding of fuel by the electroless process. The thickness of the Ni coated layer is estimated to be about 6 μm. A pre-oxidation treatment was done for the other fuel. The fuel rod was treated in the autoclave at 180 °C under 150 psi for 16 hours. The pre-oxidation thickness is estimated to be about 3 μm.



(a) **(b)**

Figure 1. SEM observation on coating layer; a) Ni coating, b) Pre-oxidation layer.

The poison of Er_2O_3 was added to the U-Mo powder prior to blending with the Al powder at a 4 wt.% content of the U-7Mo powder. The monolithic U-Mo ring with a small height was fabricated by melting and casting in a ceramic coated graphite mold. In order to have a gamma phase, heat treatment at 850℃ for 30 minutes and quenching were done. The X-ray diffraction analysis showed that the U-Mo ring structure had completely transformed to a gamma phase. Finally the ring was machined to be $^{OD}6.2\times{}^{t}0.7\times{}^{L}3.25$. The ring was inserted into the Al plug with fitting grooves and fitted with a Al fitting rod having a hole. Then the Al fitting rod was fixed by squeezing the groove sites. The diameter of the Al plug in the U-Mo ring was designed to be 0.5 mm smaller than the inner diameter of the ring. So it is expected that there will be no interaction between the U-Mo ring and Al plug. The uranium density of the monolithic U-Mo ring fuel corresponds to be the about 6 g-U/cc in the case of considering the middle cross section only. In order to eliminate an improper bond in the cladding extrusion the cladding extruding temperature was raised to 520 ℃.

2-2. Calculations of linear power, temperature, and burnup for the 2nd irradiation test fuels.

The maximum linear powers of the fuels were calculated and are represented in Table 3. The highest linear power in the dispersion fuels appeared in the U_3Si dispersion fuel. As a relatively high linear power group, the Ni coated fuel, pre-oxidation treated fuel, and standard U-7Mo could be classified. In calculating the centerline temperature thermal conductivities of the U-Mo fuels were derived from the out-core measured results of the fuel meats for the Mo contents and uranium densities.

The centerline temperatures of all the fuels at the beginning of life are calculated as in Table 3. The highest temperature in the dispersion fuels appeared in the U_3Si dispersion fuel. The reduced fuel rods of U-9Mo and U-7Mo with a higher uranium loading of 4.5 g-U and the standard U-7Mo fuels with surface treatments of Ni coating and pre-oxidation are considered to have a relatively high temperature. The temperature of the standard U-7Mo dispersion fuel of 4.0 g-U/cc is classified as medium class. The temperature of the monolithic U-Mo ring fuel was calculated to rise up to 302.9 ℃. Average burnup of the 2nd irradiation test fuels is about 35.7 at%-U235 based on the fuel management calculation as of September 4, 2003.

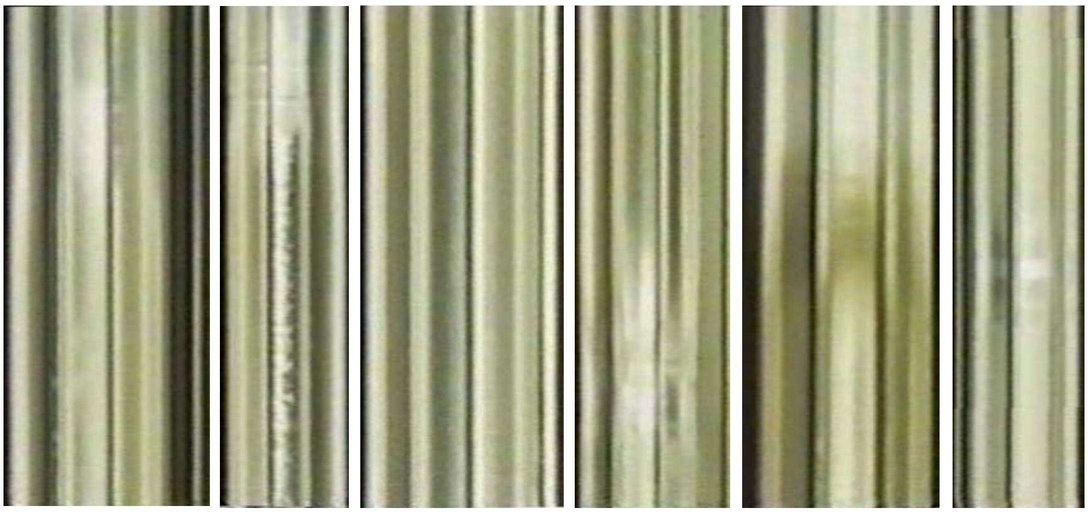
Table 3. Calculated linear powers and centerline temperatures for the fuels under the 2nd irradiation test

Fuel				Linear power (kW/m)	Centerline temperature (℃, BOL)
Material	Remarks	Loading (g-U/cc)	Diameter (mm)		
U-7Mo	High density	4.5	5.49	98.63	189.4
U-9Mo	High Mo	4.5	5.49	102.99	195.9
U-7Mo	Low density	4.0	6.35	104.37	183.8
U-7Mo	Larger particles	4.0	6.35	97.95	175.0
U-7Mo	Smaller Particles	4.0	6.35	98.47	175.7
U_3Si	Reference	4.0	6.35	112.09	201.3
U-7Mo	Poison	4.5	5.49	92.57	180.2
U-7Mo	Ring	Pure	$^{OD}6.2\times{}^{t}0.7$	150.32	302.9
U-7Mo	Ni coating	4.0	6.35	107.84	188.6
U-7Mo	Pre-oxidation	4.0	6.35	108.03	188.8

2-4. Observations on the fuels under the 2nd irradiation test

In the optical observation using the immersion camera any tangible swelling was not been detected in the irradiation fuels. Some fuels indicated a cladding surface color change into a little darker color. The most darkened fuel as in figure 2 was found to be the U_3Si dispersion fuel of a 6.35 mm standard diameter with a uranium loading density of 4.0 g-U/cc. It is assumed that the color change would be induced by the formation of aluminum hydroxide at a higher temperature. This result seems to agree with the calculation results showing the highest power rate of the U_3Si dispersion fuel. The U-Mo dispersion fuel of a 6.35 mm standard diameter with a uranium loading density of 4.0 g-U/cc had a color change as shown in figure 2.

A little luster was observed on the darkened surface of the fuel cladding. A corrosion reaction seems to happen by the formation of a dense aluminum hydroxide. These kinds of corrosion have been observed on normal HANARO driving fuels. The color of the above fuels seems to be changed more than on the driving fuels in HANARO. It is considered that the irradiation performances of the fuels would be acceptable. The U-Mo dispersion fuels of 5.49 mm diameter with the uranium density of 4.5 g-U/cc maintained a sound surface. The fuel, which is composed of U-9%Mo, was observed to have almost the same surface condition as the fuels of U-7%Mo.



(a) (b) (c) (d) (e) (f)

Figure 2. Optical observations using an immersion camera; (a) U3Si dispersion fuel, (b) standard U-7Mo dispersion fuel of 4.0 g-U/cc, (c) reduced U-9Mo dispersion fuel of 4.5 g-U/cc, (d) Ni coated fuel, (e) Pre-oxidation fuel, (f) Monolithic U-Mo ring fuel.

For the two different particle size fuels with smaller particles and larger particles it was difficult to compare the irradiation performance in the same surface observation. The fuel, on which a surface treatment of a Ni coating was done to prevent the corrosion with cooling water, revealed a good surface condition was maintained on the whole in spite of the higher linear power than the standard U-Mo fuel with a uranium loading density of 4.0 g-U/cc as in figure 2. The pre-oxidation treated fuel showed no corrosion but indications of a little grayed color on all the surface area induced by pre-oxidation. On the fuel including the poison of 4 wt.% Er_2O_3 there was not any color change.

3. The progress of the monolithic U-Mo fabrication technology development

3-1. Monolithic U-Mo thin tube

The development for the fabrication of the U-Mo tube has been carried out utilizing the continuous casting equipment, which was installed for casting uranium metal converted from spent oxide nuclear fuel. A uranium tube of an outer diameter of 10 mm and thickness of 2 mm was fabricated successfully as shown figure 3. The efforts for a thinner tube are being made. The target thickness and diameter are less than 1 mm and 7 mm, respectively. The larger diameter can be achieved easily by changing the mold size.

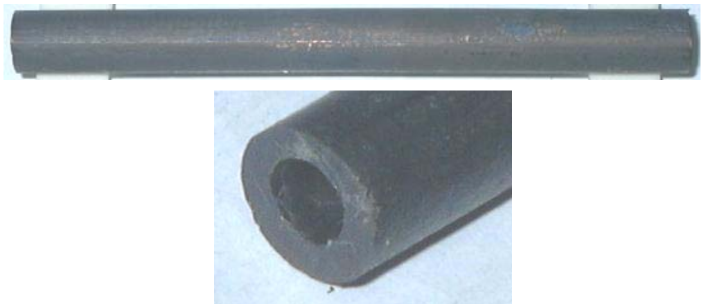


Figure 3. Uranium tube fabricated using continuous casting equipment

In the case of the unsuccessful production of a U-Mo tube by the continuous casting process, a ring type fuel would be an alternative. The dimension of the U-Mo ring will be 0.7 mm thickness, about 6.35 mm diameter, and about 10 mm height. It is considered that such a ring can be produced by investment casting process without difficulties. However, the large amount of waste and the low yield are concerns. Anyway an investment casting experiment will be done in the near future.

3-2. Monolithic U-Mo foil

The development of uranium foil fabrication technology was launched with the target of Mo-99 production. Roll casting equipment was designed and installed in 2002. The uranium foil of less than 150 μm and 50 mm width could be produced. Recently U-7wt.%Mo foil was produced successfully in the same dimension using the above equipment. The surface of the U-Mo foil exhibited a little roughness on the free-side and was fairly smooth on the roll side. The roughness test represented that Ra and Rmax are 1.99 μm and 13.24 μm for the free-side surface. On the other hand, the Ra and Rmax of the roll-side surface are 0.26 μm and 3.07 μm, respectively. U-Mo foil seems to be stronger than the pure uranium foil. The pure uranium foil with a thickness less than 150 μm is so flexible that it can be easily coiled. However, the U-Mo foil with the thickness larger than 100 μm is too stiff to be coiled. The X-ray diffraction analysis of the U-Mo foil showed the typical peaks of a gamma phase as in figure 4. The grain size of the U-Mo foil appeared to be fine at about 10 μm as shown in figure 5.

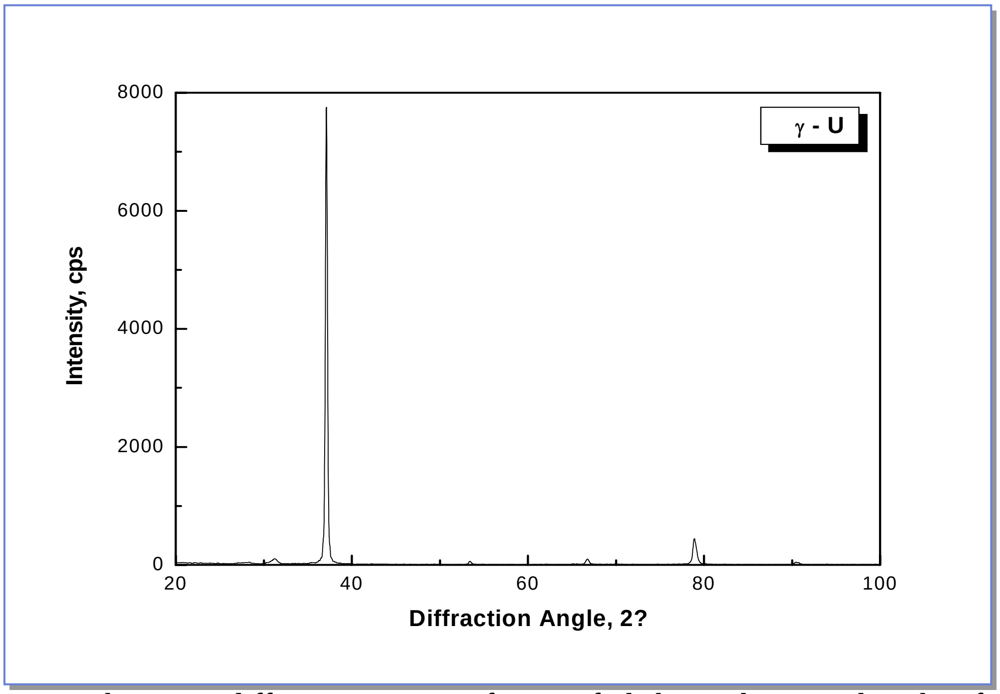


Figure 4. The X-ray diffraction pattern of U-Mo foil shows the typical peaks of gamma phase.

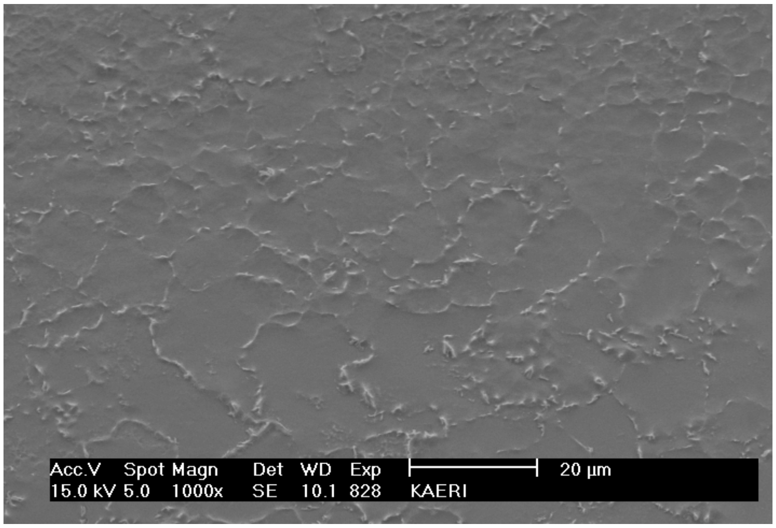


Figure 5. SEM observation on the cross section of U-Mo foil.

Presumably there would be a possibility of sheet texture formation in the foil because the foil is produced by solidification from the bottom side by contacting the cooling roll. Several pole figures by neutron diffraction have been measured. The results showed that the preferred orientation densities in foil are very small as shown in figure 6. It is considered that the detrimental effect of the preferred orientation in the foil would be almost negligible

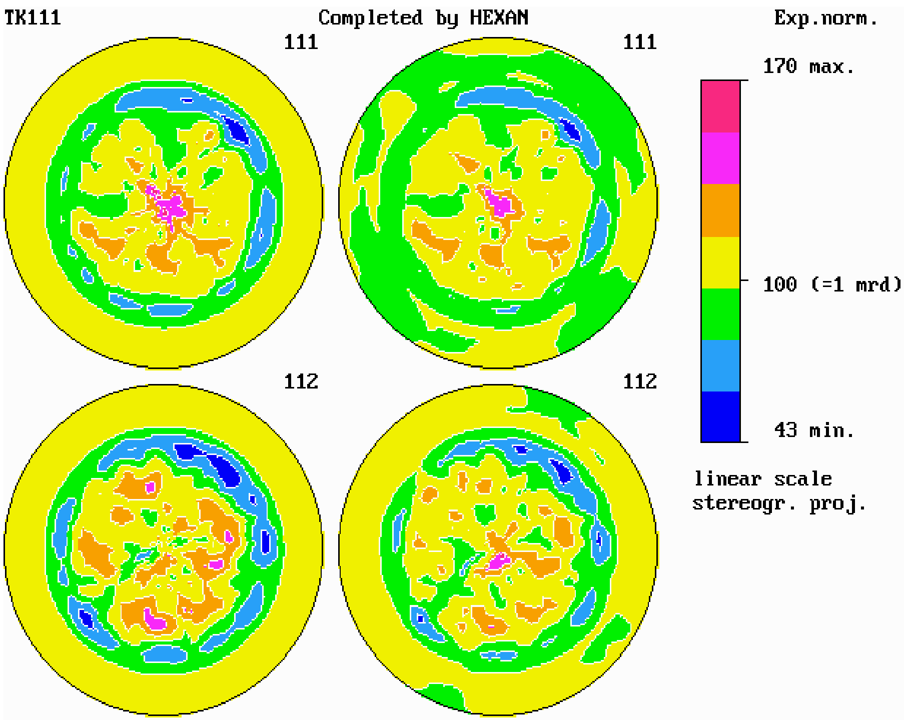


Figure 6. Pole figure taken by neutron diffraction for U-Mo foil.

4. The activities on the surface treatment of fuel cladding

4-1. Multi-layer coating of Ni and Cu

Ni coating on Al cladding is known to be exfoliated during irradiation test. One of the reasons would be imperfect adhesion or bonding between the Al cladding and the coated Ni layer. A interlayer having a good bonding with Al and Ni would improve the anti-corrosion effect. A Cu interlayer was selected as a candidate. Preliminarily a copper coating was done in less than 5 μm thickness using electroless plating process and then nickel coating was done in less than a 5 μm thickness in the same way. Some bonding tests are under way for the establishment of the optimum conditions of the coating.

4-2. Zr coating on Al cladding surface by sputtering

The above double layer coating is considered to be thick. From the aspect of the reactor performance, it was thought that a thin zirconium coating would be an alternative way. So zirconium coating was tried by sputtering. Recently zirconium coating of about 1 μm could be done without any difficulties. In the near future some bonding tests for zirconium coating are scheduled. Through the bonding tests and the optimization of Zr sputtering coating, Zr coated fuel by sputtering will be loaded in to the next irradiation test.

5. Future plan

A qualification test of the U-Mo dispersion rod fuel for HANARO will be aimed at mainly in the next irradiation test. The uranium density of the fuel rods will be basically same as in the 2nd irradiation. The PIE results of the 2nd irradiation test should be fed back to the fuel fabrication of the next irradiation test. It is expected that the 2nd U-Mo irradiation test will be terminated next February. Accordingly the next irradiation test will be possible around the end of 2004.

In addition, If a monolithic U-Mo ring fuel shows an acceptable performance in the 2nd irradiation test, a monolithic U-Mo fuel having a tube or rings will be included in the next irradiation test. The poison addition to the fuel meat can extend the fuel cycle extension so that the reduction of the spent fuel can occur. Through economic evaluation, a poison added fuel will probably be loaded. The surface-treated fuels such as multiplayer electroless coating and zirconium coating by sputtering will be loaded according to the bonding test and the optimizing results of the coating process.

6. Summary

The U-Mo dispersion rod fuels under the 2nd irradiation test were observed to be in good condition so far. The Ni coated fuel and the pre-oxidation treated fuel are maintaining the initial surface without any exfoliation of the coated layer. It is expected that the Ni coating layer could maintain a sound condition until the termination of the irradiation. The monolithic U-Mo ring fuel has not swelled by a detectable amount. The other fuels seem to be behaving in an acceptable manner under the irradiation test.

For the activities of monolithic U-Mo fuel fabrication development, some achievements such as preliminary U-Mo tube production by a continuous casting process and a successful U-Mo foil production using the roll casting process have been obtained.

In addition, some investigations on the surface-treatment of a multiplayer coating and Zr sputtering coating has revealed the possibility of eliminating the problem of a temperature rise due to corrosion layer formation having a quite low conductivity.

The next irradiation test will aim mainly at the qualification of the U-Mo dispersion fuel for HANARO around the end of next year. In the irradiation fuel buddle, some fuels related to the basic investigation tests of the monolithic U-Mo fuel and the surface-treatment for anticorrosion will be loaded.

7. Acknowledgement

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8. Reference

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